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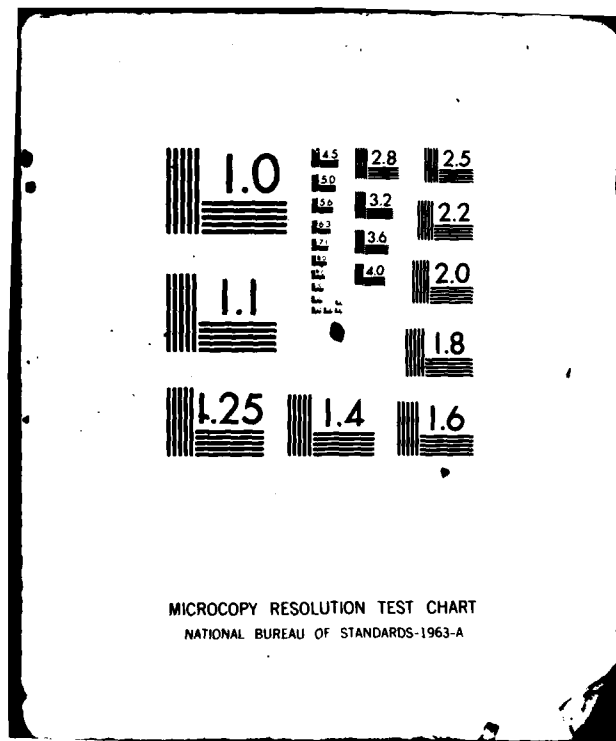
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**MAGNETOSTRICTION AND
CHARACTERIZATION OF BRIDGMAN
GROWN RARE-EARTH IRON ALLOYS**

BY H. T. SAVAGE,
A. E. CLARK

RESEARCH AND TECHNOLOGY DEPARTMENT

1 NOVEMBER 1981

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20. ABSTRACT (Continue on reverse side if necessary and identify by block number) We have prepared large samples of Tb. ₂₇ Dy. ₇₃ Fe ₂ and Tb. ₂ Ho. ₅₈ Dy. ₂₂ Fe ₂ by a Bridgman-like technique developed at United Technology. The samples have much larger values of saturation magnetostriction than vertically zoned materials. The materials have been characterized by metallographic and x-ray topography studies. Seeded growth resulted in a superior crystallo- graphic orientation. Plane front growth was achieved over most of the boule.		

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FOREWORD

The magnetic and magnetomechanical properties reported here are developments of a program to develop magnetostrictive materials for high power sonar. The materials are an alloy of terbium-iron and dysprosium-iron Laves phase compounds. The magnetoelastic strains are very large and very anisotropic.

Results of the second year effort preparing large sample crystals of the highly magnetostrictive Tb compounds utilizing high temperature-gradient furnaces are reported here. This method has been identified as most promising to yield large single crystals (3" x ½" dia.) for sonar transducer elements. Plane front growth was achieved over the entire length in one crystal for the first time. Seeded growth was achieved for the first time. Part of the results will be published in the proceedings of the 1982 Durham conference on the rare-earths and actinides.

This study was carried out in the Solid State Branch of the Radiation Division. The materials development was sponsored by the NRL Material Program under the direction of Howard Lessoff. Magnetic measurements and fabrication of prototype transducer components utilizing these materials is being carried out under the sponsorship of the NOSC Sonar Transducer Sciences Program. Research on the magnetoelastic properties of highly magnetostrictive rare earths is sponsored by the Office of Naval Research and the NSWC Independent Research Fund.

Ira M. Blatstein
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CHAPTER 1

INTRODUCTION

The cubic Laves-phase rare-earth Fe_2 compounds have huge values of magnetostriction constants (λ) and anisotropy energy (E_k). However, with an appropriate alloying of binary compounds, ternaries of the form $\text{R}_x\text{R}_{1-x}\text{Fe}_2$ may be obtained with large values of λ but with E_k reduced by two orders of magnitude.¹ Large values of the magneto-mechanical coupling factor, k_{33} , have been observed in $\text{Tb}_{.27}\text{Dy}_{.73}\text{Fe}_2$ (Terfenol). The magnetostriction is quite anisotropic with $\lambda_{100} \ll \lambda_{111} = 1.6 \times 10^{-3}$. Because the large, anisotropic magnetostriction introduces large inhomogeneous strains in random polycrystals (RPC), the permeability is quite low.² This restricts magneto-mechanical activity.

We previously reported efforts in grain orienting to enhance magnetomechanical activity.³ Bridgman growth and horizontal zoning methods were examined during FY78. During FY79, methods were developed to prepare high coupling oriented samples by a vertical zoning technique.⁴ This material is prepared by passing a vertical molten zone (VMZ) along a vertical rod. We achieved considerable improvement with this over polycrystalline material. Coupling factors larger than .7 were achieved in long rods. Random polycrystalline materials have values of .55. Raytheon Corporation is now building a transducer using long rods of vertically zoned material.

Last year's report⁵ covered our first extensive attempt to obtain single crystal material in a manner suitable for large scale production of transducer rods. The method has been developed by United Technology for single crystal turbine blades. We wish to adapt it to the production of single crystal $\text{Tb}_{.27}\text{Dy}_{.73}\text{Fe}_2$. The method is basically a Bridgman growth technique. A boron nitride crucible is lowered through a temperature gradient. Since a large temperature gradient is necessary, the upper part of the crucible is held well above the melting point (called "super-heat"). The ampule can be lowered into liquid to enhance the temperature gradient.

¹A. E. Clark, AIP Conf. Proc. 18, 1015 (1974).

²H. T. Savage, A. E. Clark, and J. M. Powers, IEEE Trans. on Magnetics, MAG-11, 1355 (1975).

³H. T. Savage, R. Abbundi, A. E. Clark, "Permeability, Magnetomechanical Coupling and Magnetostriction in Grain-Oriented Rare Earth-Iron Alloys," NSWC/WOL TR 78-197.

⁴H. T. Savage, R. Abbundi, A. E. Clark, "Magnetomechanical Coupling and Magnetostriction in Vertically Zoned $\text{Tb}_{.27}\text{Dy}_{.73}\text{Fe}_2$," NSWC TR 79-463.

⁵H. T. Savage, R. Abbundi, A. E. Clark, "Magnetostriction and Characterization of Bridgman Grown Rare-Earth Iron Alloys," NSWC TR 81-29.

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Last year we had considerable trouble with crucible interation and poor orientation. We have reduced the crucible problem and had partial success with seeded growth for proper orientation. X-ray topographical analysis has been started.

CHAPTER 2

SUMMARY OF RESULTS

The previous year's work consisted of growing several large crystals.⁵ Plane front growth was not achieved over the entire length of the crystal. Considerable difficulty was encountered in the interaction of the crucible with the melt. In general, large amounts of Widmanstatten precipitate (WSP) were encountered. Magnetostriction was measured. This year we have achieved the following results:

1) We made 10 runs with three successes, crystal Nos. 5, 8 and 10. Improvement in the structural integrity of the crucible was a factor.

2) Achieved plane front growth over the entire length in No. 10 for the first time.

3) We had partial success in seeded growth. We have had a poor orientation in the past. We want a $\langle 111 \rangle$ growth direction since this yields maximum magnetostriction. Crystal No. 5, which had a $\langle 100 \rangle$ growth direction, was cut into seeds. Seeded growth in crystals 8 and 10 yielded a growth direction about 10° from $\langle 111 \rangle$, a considerable improvement. A $\langle 111 \rangle$ section is being cut from No. 8 which is long enough to simulate a transducer rod.

4) We have begun using x-ray topography in addition to our other methods to characterize our crystals. It is the best overall test for crystalline perfection. We have seen inhomogeneous strain fields due to both WSP and magnetic domains.

⁵See footnote 5 on page 7.

CHAPTER 3

TERFENOL-D SINGLE CRYSTAL PREPARATION

The major objective of this program is to achieve large (3" x.5" dia.) single crystal transduction elements of $\text{Tb}_{.27}\text{Dy}_{.73}\text{Fe}_{1.95}$ (Terfenol-D). Ten samples were grown. (See Table 1.) The first four attempts were unsuccessful. Pyrolytic BN crucibles, purchased from Union Carbide, were defective, causing leaking in penetration of the melt through the BN layers. In the first three runs, the melt did not completely penetrate the crucible, making it difficult to determine the cause of the failures. In the fourth case, the melt penetrated the entire crucible wall, revealing the cause of the previous failures.

The fifth run was the first success. The starting boule (3" x.5" dia.) contained high purity $\text{Tb}_{.27}\text{Dy}_{.73}\text{Fe}_{1.95}$ from Ames Laboratory, Iowa State University. As in all except the final run, a tapered BN crucible was fitted with a hot pressed BN tip to hold the seed. The seed was a 1.3 cm long $\langle 111 \rangle$ sample of $\text{Tb}_{.27}\text{Dy}_{.73}\text{Fe}_{1.95}$ spark-cut from an earlier run. Directional growth commenced at 1 cm/hr for a 1 cm distance. This was followed by a linear transient growth from 1 to 10 cm/hr for the final 3.5 cm. To minimize crucible interaction, a maximum temperature of 1350° in the melt was held at 200 Torr argon pressure during growth. The tin bath was 265°C. The resulting crystal, Figure 1, was a single grain oriented 40° from $\langle 111 \rangle$. It was hypothesized that the incorrect orientation was a result of complete meltback of the seed. Unlike the earlier attempts, Run #5 possessed a relatively small amount of Widmanstätten precipitate. This can be seen in the micrograph of Figure 2. A comparable micrograph for Run #1 is shown in Figure 3. Samples from Run #5 were used for STEM and x-ray diffraction topographical analyses.

Run #8 was the second success (see Figures 4 and 5). A large single crystal was grown, however again not aligned along $\langle 111 \rangle$, even though satisfactory seed meltback occurred. A large single crystal was obtained but aligned 8° from $\langle 111 \rangle$. This failure to propagate the seed direction was surprising and runs #9 and #10 were carried out to allow close examination of the seed-tapered sample interface. The first long single crystal for magnetostrictive studies will be obtained from this run.

Run #9 yielded the second crystal closely, but not exactly aligned along $\langle 111 \rangle$. However, it fractured upon removal. Run #10 yielded the third consecutive large single crystal. As expected, there was no close alignment with the $\langle 111 \rangle$ seed. Examination of the seed and seed-sample interface showed the presence of a region of dendritic growth. The absence of plane front growth in this critical region reveals the existence of a low temperature gradient in the tapered part of the composite RN assembly. A thinning of the wall at the junction of the seed and the tapered sample should eliminate the low temperature gradient and maintain the seed orientation. This change will be implemented in the runs carried out in the next phase of this program.

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In summary, four large single crystals were grown under this program, three consecutively. Two closely aligned along the 111 direction (8° away). A .22" dia. specimen has been prepared from Run #8 for magnetostriction and d-constant measurements.

TABLE 1 Tb_{0.27}Dy_{0.73}Fe_{1.95} CRYSTAL GROWTH EXPERIMENTS

<u>Run No.</u>	<u>Growth Velocity</u> (cm/hr)	<u>Crucible</u>	<u>Seed</u>	<u>Remarks</u>
1	1-10	Pyrolytic BN	<111> Fe	Polycrystal
2	1-10	HBR-BN	<111> Fe	Seed did not melt, reaction, polycrystal
3	1-10	HBR-BN	<111> Fe	Seed melted back, reaction, polycrystal
4	1-10	Pyrolytic BN +HBR tip	<111> Fe	Seed melt back, oxidized, polycrystal
5	1-10	"	<111> R Fe ₂	Large single grain 12° - 14° from <001>
6	1-10	"	<111> R Fe ₂	Seed left out inadvertently
7	1-10	"	"	No seed melt back, polycrystal
8	1-10	"	"	Seed melt back, single grain 8° - 12° from <111>
9	1-10	"	"	Single grain fractured in removal, 8° - 12° from <111>
10	1-10	"	"	Single grain, approximately 40° from <111>

CHAPTER 4

CHARACTERIZATION

This year we initiated an x-ray diffraction topography structure study of our materials. Topography is a unique tool in the sense that we can see microscopic strain fields with very low magnification. Large areas (25 cm^2) can be seen with a single exposure. Resolution can be $2 \mu\text{m}$. Interpretation is direct, since we basically see the inhomogeneous strain fields caused by the defect structure. Figure 6 is a scanning Lang topograph of a piece of crystal 5. There would be no contrast if No. 5 were a perfect crystal. WSP causes inhomogeneous strains which alter the Bragg conditions. There is an additional benefit from using topography. We can see domain structure with topography since the magnetostriction disturbs the Bragg conditions. Figure 7 is a reflection topograph of a piece of crystal 5. The vertical and some horizontal structure is caused by domains. In the future we hope to see the interaction of the domain wall with defects. The strength of the interaction of the domain wall with defects. The strength of the interaction will tell us whether the defect is important in the magnetization process (and therefore in the transduction process).

Figure 8 is a metallograph of a piece of crystal 5. The WSP is not uniform but appears only in certain regions (the darker regions). The pattern is indicative of constitutional supercooling (CS). This is the first time we have seen CS. The effect occurs because of a build-up of Fe in the liquid at the interface. If the explanation of the observed pattern of WSP is CS then we may be able to prevent the formation of WSP by altering the composition. Preventing WSP is better than removing by heat treatment as we do now.

CHAPTER 5
MAGNETOSTRICTIVE RIBBONS

We have reported the largest magnetomechanical coupling ever observed⁶ in fiscal 1981. A subsequent publication⁷ in fiscal 82, using a different technique, supports the previous work. This publication also provides the necessary data and theory for the design of accelerometers, robotic sensors and sonar hydrophones. The ribbons can now be considered the most sensitive of material available for sensors of this type. Figure 9 shows the coupling factor as a function of field. Figure 10 shows the inverse susceptibility versus tension. This figure contains most of the information needed for device design. We have a theory for the magnetization process that allows us to predict device performance. We have constructed a furnace that anneals toroidal shaped samples. This geometry is necessary for some devices. The ribbon materials are ready now for some applications.

⁶C. Modzelewski, H. T. Savage, L. T. Kabacoff, and A. E. Clark, "Magnetomechanical Coupling and Permeability in Transversely Annealed Metglas 2605 Alloys," IEEE, Trans. on Mag., MAG-17, 1981, p. 2837.

⁷M. Spano, K. Hathaway, and H. T. Savage, "Magnetostriction and Magnetic Anisotropy of Field Annealed Metglas 2605 Alloys Via dc M-H Loop Measurements Under Stress," Journal of Applied Physics, to be published.

CHAPTER 6

RECOMMENDATIONS

The goal of this program is to develop mass production techniques to inexpensively produce large samples of highly magnetostrictive rare-earth iron alloys. We want magnetostrictions in excess of 10^{-3} at 500 Oe. High quality single crystals offer the largest possible strains at low fields. To obtain high quality crystals, we recommend the following:

- 1) Continued x-ray topography analysis of the crystals for defect analysis.
- 2) Variation of the starting composition to eliminate Widmanstatten precipitate initially instead of by heat treatment.
- 3) Continued seeded growth attempts. The reason for the transient appearance of the second phase must be found and the growth procedure altered to improve the orientation.
- 4) The ribbon materials are now ready for the construction of an accelerometer. We intend to construct such a device on a ceramic substrate next year. The device will be designed as closely as possible to have the necessary characteristics for a customer at NSWC.



FIGURE 1 CRYSTAL NO. 5

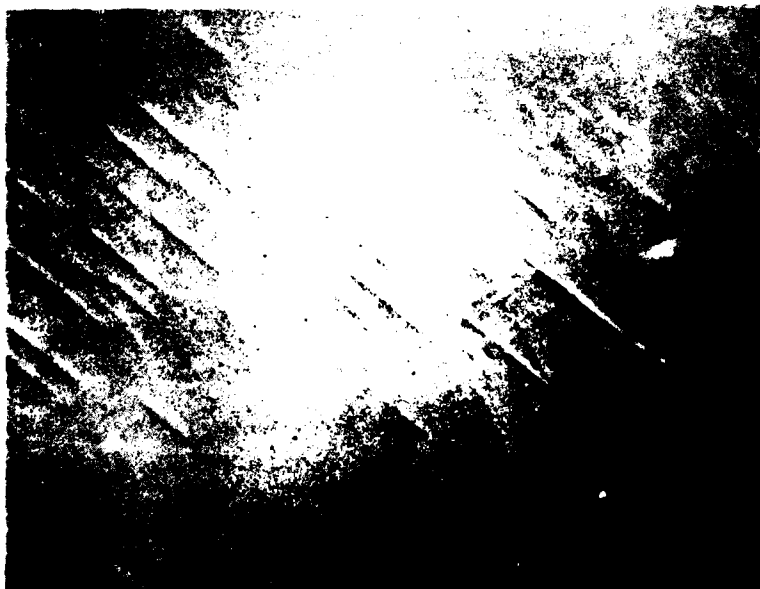


FIGURE 2 MICROGRAPH OF A PIECE OF CRYSTAL NO. 5

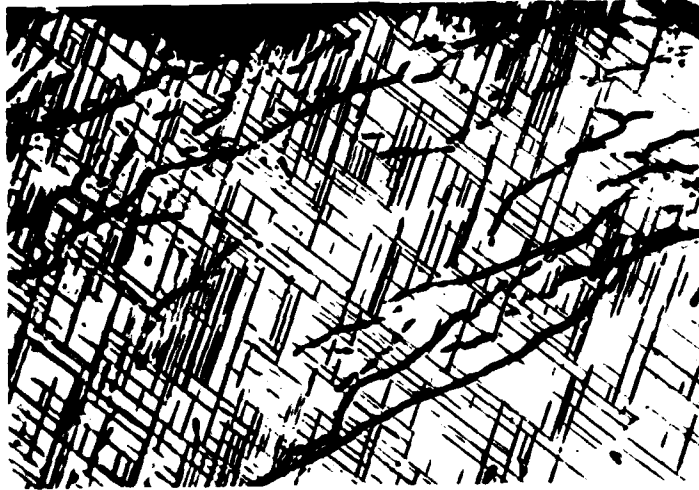


FIGURE 3 MICROGRAPH OF A PIECE OF CRYSTAL NO. 1
SHOWING CONSIDERABLY MORE WIDMANNSTATTEN
PRECIPITATE THAN IN FIGURE 2

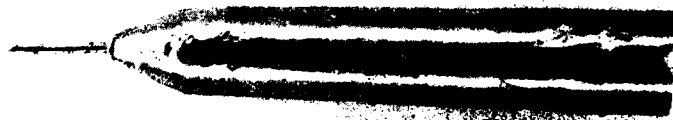


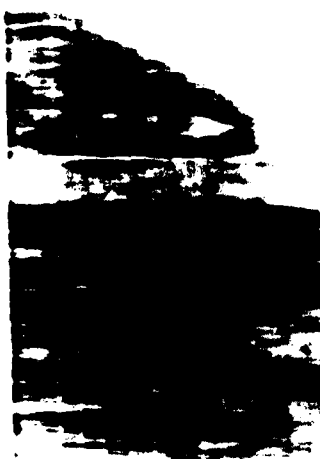
FIGURE 4 CRYSTAL NO. 8. THE PROTRUSION IS THE SEED.



FIGURE 5 LAUE PATTERN OF CRYSTAL NO. 8. THE OFF-AXIS $\langle 111 \rangle$ SYMMETRY IS EVIDENT.

H0 Plane

Magnification = x 5



$\rightarrow g = 440$

$\{ \text{Mo radiation (K}\alpha_1) \}$

$\langle 110 \rangle$

\uparrow
 $\rightarrow 100$

FIGURE 6 SCANNING LANG REFLECTION TOPOGRAPH OF A PIECE OF CRYSTAL NO. 5. THE CONTRAST IS CAUSED BY THE INHOMOGENEOUS STRAINS.

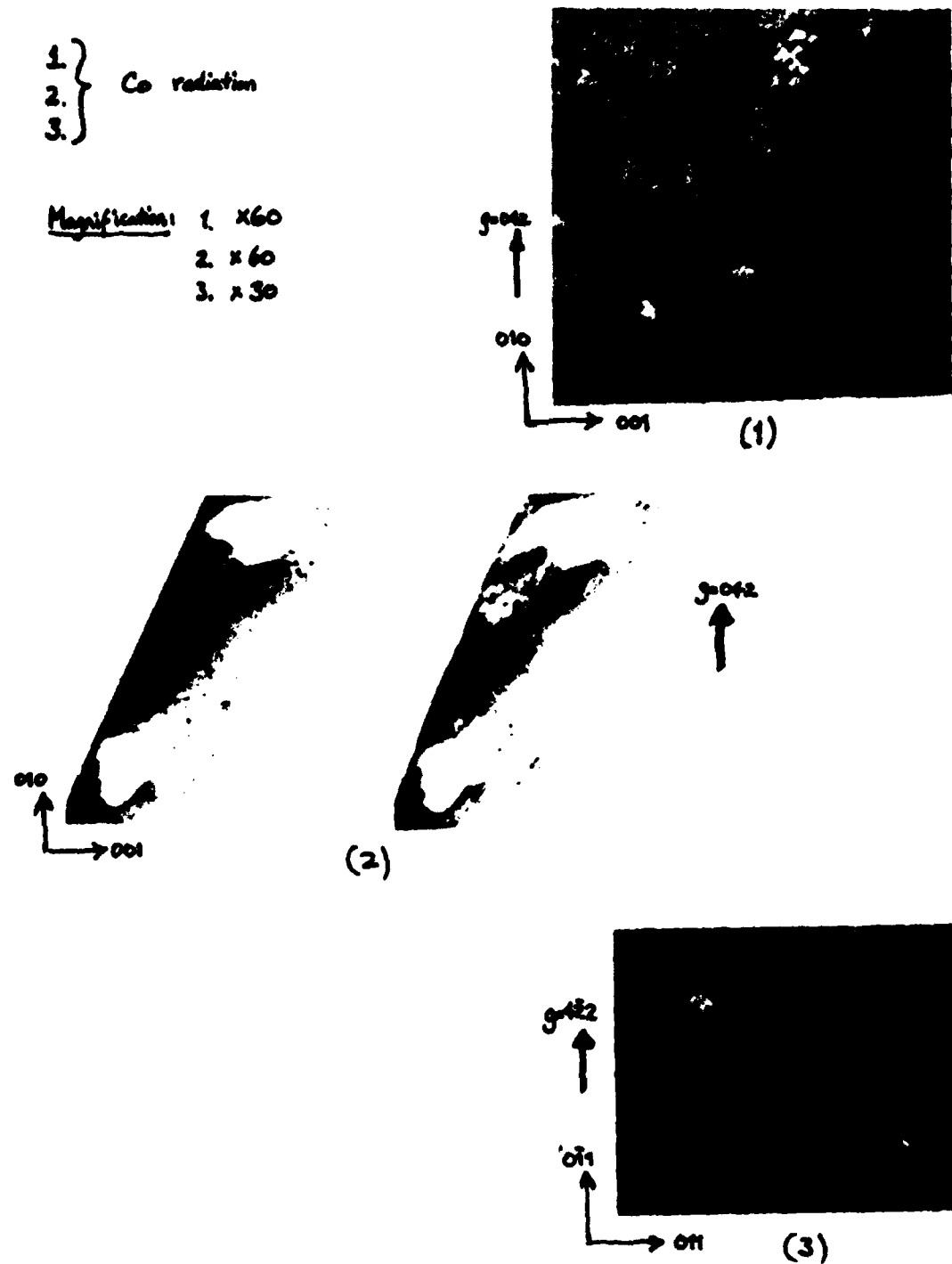


FIGURE 7 X-RAY REFLECTION BERG-BARRET TOPOGRAPH OF A PIECE OF CRYSTAL NO. 5. THE FAINT VERTICAL AND HORIZONTAL LINES ARE CAUSED BY DOMAIN STRUCTURE.

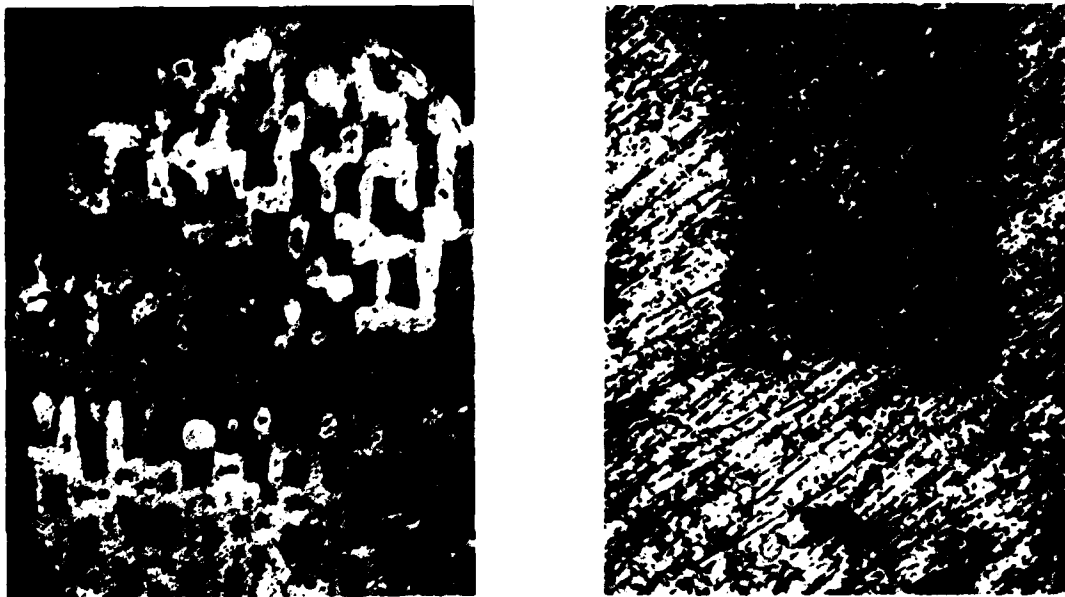


FIGURE 8 METALLOGRAPH OF CRYSTAL NO. 5.
THE DARKER REGIONS CONTAIN WSP
WHILE THE LIGHTER ONES DO NOT.

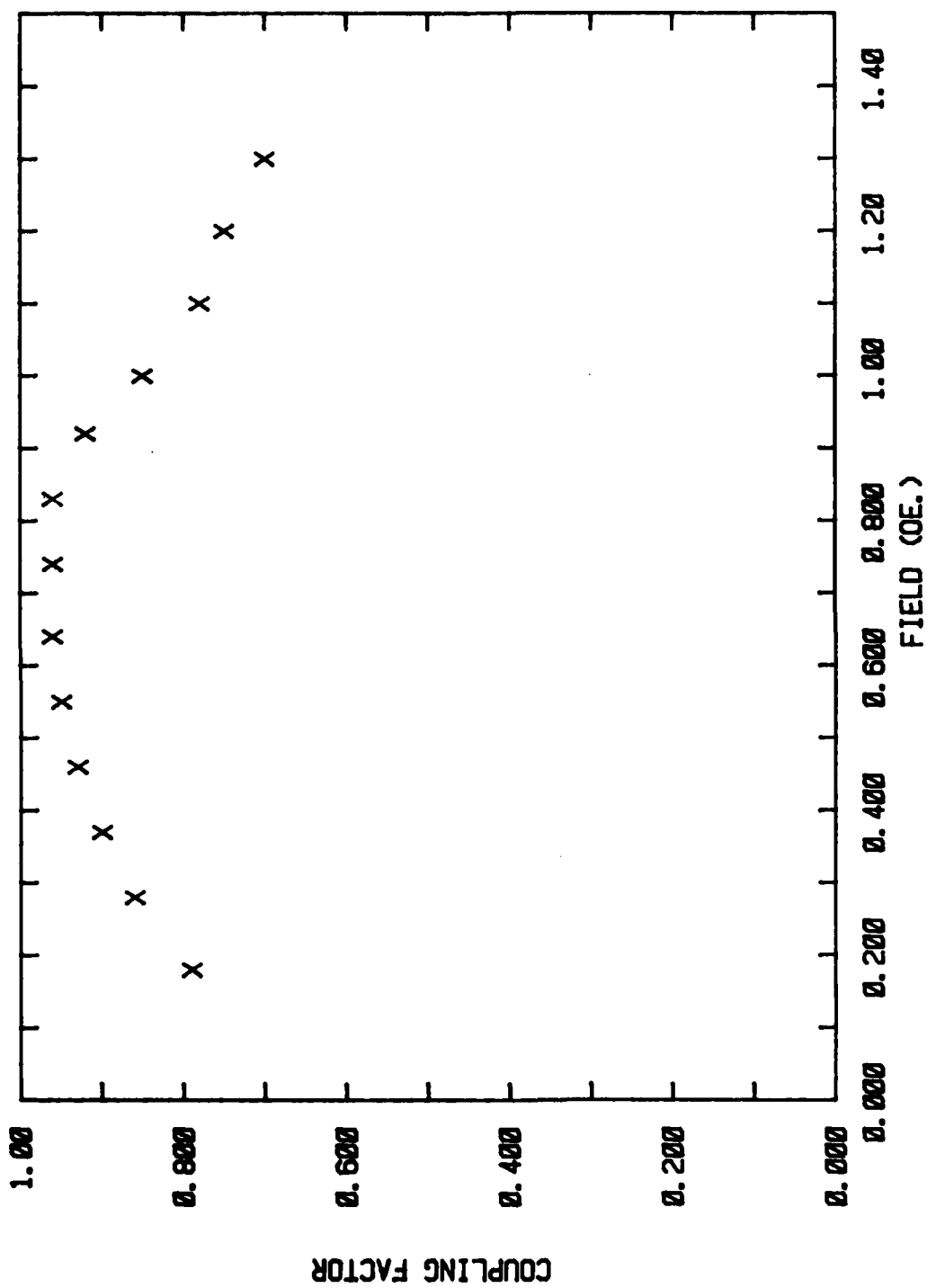


FIGURE 9 COUPLING FACTOR VS. BIAS FIELD IN METGLAS 2605SC

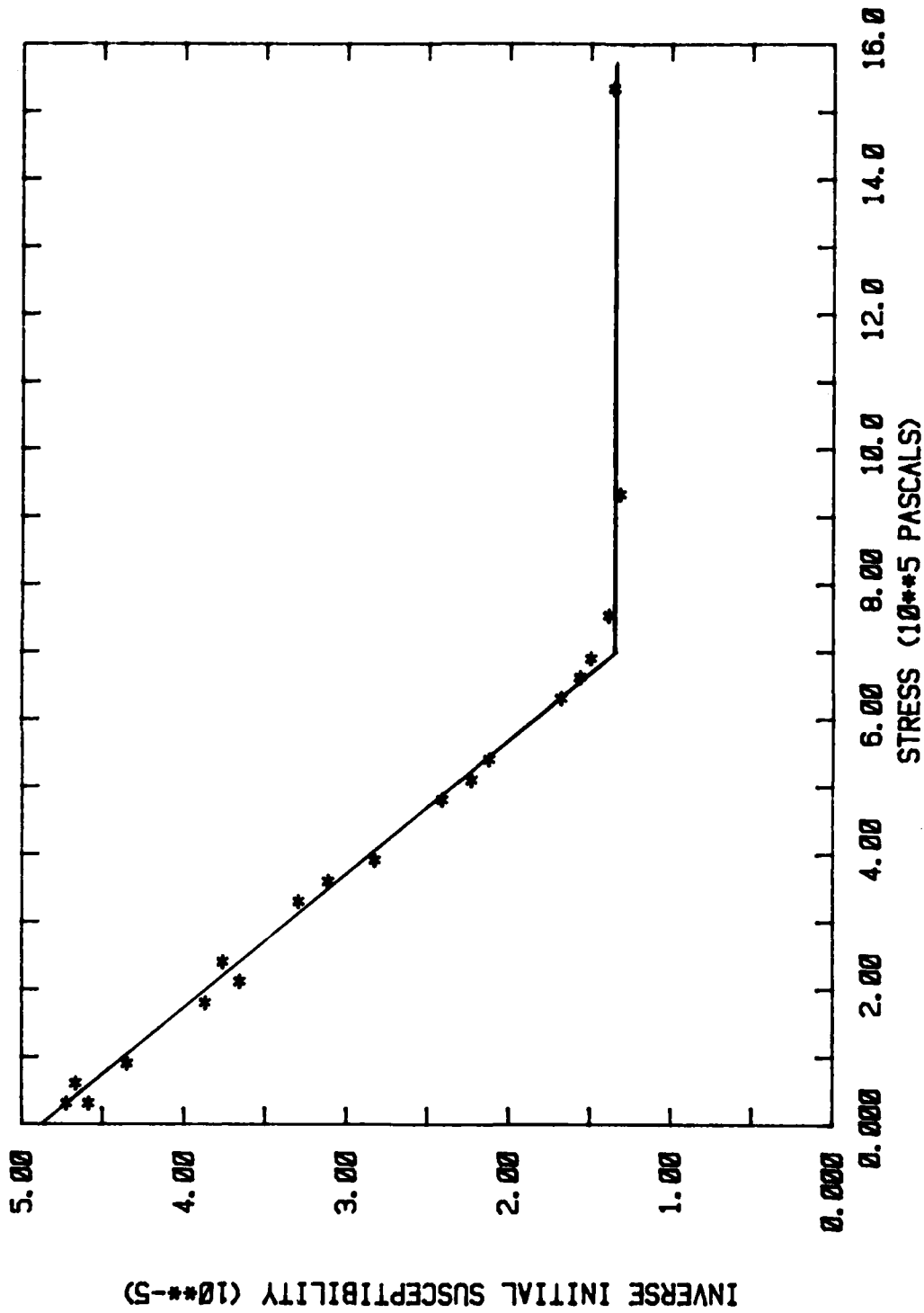


FIGURE 10 INVERSE SUSCEPTIBILITY VS. APPLIED STRESS IN METGLAS 2605SC

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